

INDEFINITENESS REJECTION

The Examiner has stated that the "molecular weight limitation renders claim 5 indefinite because it is not specified how the molecular weight is determined". The difference between "wt. average MW" and "average MW" is well known to someone of ordinary skill in polymer science.

Applicants have previously submitted that the molecular weight of the copolymers used in the invention are "weight average molecular weights". Generally, molecular weights of polymers are discussed in G.M. Barrow, "Physical Chemistry", Second edition, McGraw-Hill Book Company, 1966, pages 789-810 in the chapter "MACROMOLECULES IN SOLUTION". In particular, the determination of weight average molecular weights by light scattering is described on pages 798-805. A copy of these pages are submitted with this response for the Examiner's review. The Applicants' have determined the weight average molecular weight of the copolymers used in the invention by light scattering.

Accordingly, Applicants' believe the Examiner's § 112 rejections have been overcome and should be withdrawn.

PRIOR ART REJECTIONS

The pending claims stand rejected under § 103(a) as being unpatentable over Smigo et al., alone, or in view of Oliver et al. or Kobayashi et al.; and as being unpatentable over Kono et al. or Kashiwazaki et al. Applicants respectfully disagree.

Independent Claim 12 has been amended to particularly define the recording sheet layer of the invention as including at least one copolymer and a binder or mixture of binders "wherein the quantity of said copolymer is between 10 to 75% of the combined amount of said copolymer and binder." Support for this amendment is found in Applicants examples.

In addition, new Claim 13 has been added as an independent claim defining a preferred embodiment where the binder is gelatin.

In contrast to the present invention as defined in the

amended claims, Smigo discloses a layer comprising a copolymer of vinyl alcohol and vinyl amine, having **no other binders present**.

Further, as defined in the amended claims, the amount of copolymer in the invention layer is "between 10 to 75% of the combined amount of said copolymer and binder." This combination provides an ink jet recording sheet with improved light fastness, water fastness and greater stability. None of the art cited by the Examiner, either alone or in combination teach the structure as now defined in the claims.

In addition, Applicants respectfully refer the Examiner to the previous amendments submitted wherein arguments were presented to specifically overcome each of the cited references. Applicants assert these prior arguments are now fully supported by the pending claims and overcome all the cited art.

In view of the foregoing, Applicants submit that this application is now in condition for allowance. No new matter has been introduced by this Amendment. Reconsideration of this application and allowance of Claims 3-13 are hereby requested. If a telephone interview would be useful to advance this case, then the Examiner is invited to telephone the undersigned.

Respectfully submitted,

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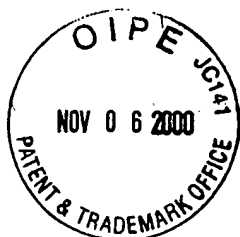
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polymer chains will be solvated and will open up; in a "poor" solvent they will tend to remain coiled up. The expectation of a high intrinsic viscosity for polymers in good solvents compared with poor solvents is borne out by the result for polystyrene of 1.20 to 1.30 in good, aromatic solvents and 0.65 to 0.75 in poor, aliphatic solvents.

Again some deductions as to the shape of polymer molecules are possible. The important use of viscosity measurements is, however, in the rapid determination of relative molecular weights.

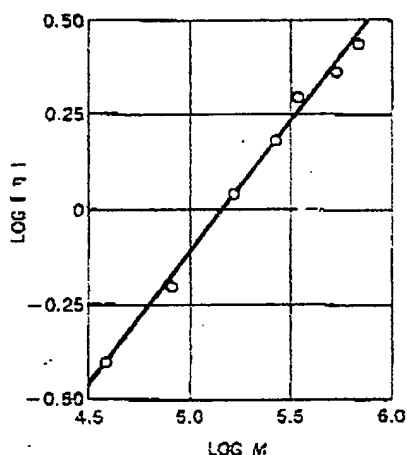
## \* 25-12. LIGHT SCATTERING

One of the most distinctive features of a colloidal, or macromolecule, solution is the scattered light, or Tyndall effect, that is observed when a light beam is passed through such a solution. This scattered light can be used in two different ways to help elucidate the nature of colloidal solutions.

The first application is made in the *ultramicroscope*, which is shown schematically in Fig. 25-9. Here the sample is observed through a microscope at right angles to the direction of the entering light beam. Each colloid particle, larger than about 10 Å diameter in very favorable cases, will produce an observable point of scattered light. The individual particles can then be counted, and if the micro-

FIGURE 25-8

Relation of  $[\eta]$  to  $M$  for polyisobutylene fractions in cyclohexane at 30° C. [From W. R. Krigbaum and P. J. Flory, *J. Am. Chem. Soc.*, 75:1775 (1953).]



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TABLE 25-7

Constants for Eq. [25] for various polymer-solvent systems\*

Polymer	Solvent	$t$ (°C)	Mol.-wt range	$K$	$a$
Polystyrene	Benzene	25	32,000-1,300,000	$1.03 \times 10^{-4}$	0.74
Polystyrene	Methyl ethyl ketone	25	2,500-1,700,000	$3.9 \times 10^{-4}$	0.58
Polyisobutylene	Cyclohexane	30	6,000-3,150,000	$2.6 \times 10^{-4}$	0.70
Polyisobutylene	Benzene	24	1,000-3,150,000	$8.3 \times 10^{-4}$	0.50
Natural rubber	Toluene	25	40,000-1,500,000	$5.0 \times 10^{-4}$	0.67

\* From P. J. Flory, "Principles of Polymer Chemistry," Cornell University Press, Ithaca, N.Y., 1953.

scope focuses on a definite, known volume of solution, the number of particles per unit volume can be determined. Such data, along with the measurable weight of macromolecule material per unit volume, lead to a value of the average mass of the individual particles.

It should be emphasized that none of the details of the particles can be observed. They merely act as scattering centers, and one observes points of light. Furthermore, unless the refractive index of the colloid particle is very different from that of the solvent, the scattered light is too weak to be seen. The similarity of the refractive index of most macromolecules to the medium in which they are dispersed means that little scattered light will be given off. The method is therefore mostly applicable to inorganic colloids.

The second application of the scattering of light depends on the measurement and interpretation of the amount of light scattered in various directions as a beam of light passes through a solution of macromolecules. In some experiments the total amount of the scattered light is deduced from the decrease in intensity of the incident beam as it passes through the sample. Just as for Beer's law for the absorption of light (Sec. 15-17), one has the relation

$$I = I_0 e^{-\tau l} \quad [26]$$

where  $\tau$  is the measure of the decrease in incident-beam intensity per unit length of a given solution and is known as the *turbidity*. In some experiments, on the other hand, the intensity of light scattered in various directions is measured directly, rather than inferred, from the decrease in intensity of the incident beam.

That the scattered light is related to the particle size and shape can now be shown. We first consider the effect of particles that are small compared with the wavelength of the radiation. Incident plane-polarized radiation imposes, as Fig. 25-10 illustrates, an electric field

$$\mathcal{E} = \mathcal{E}_0 \sin 2\pi \nu t \quad [27]$$

at the particle. If the particle has a polarizability  $\alpha$ , there will be an

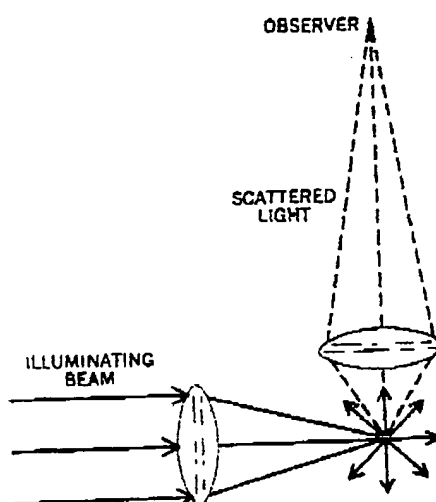


FIGURE 25-9

*The detection of particles in the ultramicroscope.*

induced-dipole moment given by

$$\begin{aligned}\mu_{\text{induced}} &= \alpha \tilde{\epsilon} \\ &= \alpha \tilde{\epsilon}_0 \sin 2\pi \nu t\end{aligned}\quad [28]$$

It is this oscillating-dipole moment that emits secondary radiation and causes the particle to be a scattering center.

It has been mentioned in connection with the difficulties of early atomic theories that in classical electromagnetic theory an accelerated charge must emit electromagnetic radiation. This result can be applied to show what secondary radiation will be emitted by the oscillating induced dipole.

The oscillating-dipole moment of Eq. [28] can be formally written as a charge of value  $\alpha \tilde{\epsilon}_0$  oscillating with a unit amplitude relative to an equal and opposite charge. Thus we can write

$$\mu_{\text{induced}} = (\alpha \tilde{\epsilon}_0)x \quad [29]$$

with

$$x = \sin 2\pi \nu t \quad [30]$$

In this way a picture is obtained of an induced charge  $\alpha \tilde{\epsilon}_0$  in the irradiated particle vibrating with simple harmonic motion. This motion, furthermore, involves an acceleration  $d^2x/dt^2$  which is calculated from Eq. [30] as

$$\frac{d^2x}{dt^2} = 4\pi^2 \nu^2 \sin 2\pi \nu t \quad [31]$$

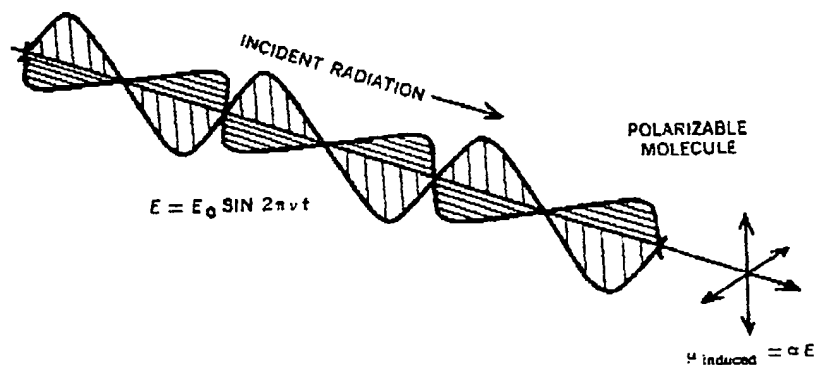
It is necessary now to quote, without derivation, the very important classical electromagnetic result that the acceleration of a charge  $q$  leads to the emission of electromagnetic radiation which produces at a distance  $r$  and angle  $\phi$ , as in Fig. 25-11, from the oscillating charge, an electric field  $e$  given by the expression

$$e = -\frac{q}{c^2} \frac{d^2x}{dt^2} \frac{\cos \phi}{r} \quad [32]$$

With this result one calculates that the radiation field of the dipole induced in the particle by one component of the incident radiation

FIGURE 25-10

The induced-dipole moments produced by the two components of incident radiation.



tion is

$$e = -\frac{\alpha\epsilon_0 4\pi^2 \nu^2 \sin 2\pi\nu t \cos \phi}{c^2 r} \quad [33]$$

The propagation of this radiation through space with a velocity  $c$  can be represented by including a sinusoidal space dependence to give

$$e = -\frac{4\pi^2 \nu^2 \epsilon_0 \sin 2\pi\nu(t - x/c) \cos \phi}{c^2 r} \quad [34]$$

It is, however, not the electric field of the radiation but rather the energy content that is of interest. This energy is directly related to the square of the field amplitude, and with this relation the intensity, or energy, of the secondary beam is calculated as

$$i = \frac{16\pi^4 \nu^4}{c^4 r^2} \alpha^2 \epsilon_0^2 \sin^2 2\pi\nu\left(t - \frac{x}{c}\right) \cos^2 \phi \quad [35]$$

What is of importance for comparison with experimental results is the intensity of scattered radiation, at various angles, compared with the intensity of the incident radiation. This incident plane-polarized radiation can be depicted as entering the sample according to the relation

$$I_0 = \epsilon_0^2 \sin^2 2\pi\nu\left(t - \frac{x}{c}\right) \quad [36]$$

and this expression can be inserted into Eq. [35] to give

$$\frac{i}{I_0} = \frac{16\pi^4 \nu^4}{c^4 r^2} \alpha^2 \cos^2 \phi \quad [37]$$

Introduction of the radiation wavelength by the relation  $\nu = c/\lambda$  then gives

$$\frac{i}{I_0} = \frac{16\pi^4}{r^2} \frac{\alpha^2 \cos^2 \phi}{\lambda^4} \quad [38]$$

When ordinary, nonpolarized radiation is used for the incident beam, the induced-dipole moment in the sample can be considered to have two mutually perpendicular components. The scattered beam consists, then, of two perpendicular components like that of Eq. [38]. This net scattered beam is related to the angle  $\theta$  of Fig.

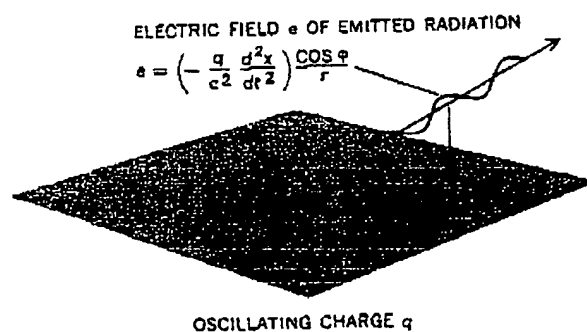


FIGURE 25-11

The angular dependence of the radiation emitted from an oscillating charge.

25-12 by the equation

$$\frac{i}{I_0} = \frac{8\pi^4 \alpha^2}{\lambda^4 r^2} (1 + \cos^2 \theta) \quad [39]$$

This angular dependence is best verified by checking that it gives the correct summation of the two plane-polarized components in various special directions.

The intensity predicted by Eq. [39] for the scattered beam from small particles is illustrated in Fig. 25-12.

It should be noticed that the forward and backward scattering are equal. Furthermore, the fourth-power dependence of the scattering on the wavelength shown by Eq. [39] should be noticed. It is, for example, to this enhanced scattering of short-wavelength radiation that the blue color of the sky is attributed. The short-wavelength blue end of the visible spectrum is scattered more than the long-wavelength red end, and the "background" color of the sky is therefore blue.

The interpretation of the scattering of radiation that has culminated in Eq. [39] can be brought to a comparison with the experimental quantity, the turbidity. For many samples the amount of scattering is small, and the turbidity expression

$$\frac{I}{I_0} = e^{-\tau l}$$

or

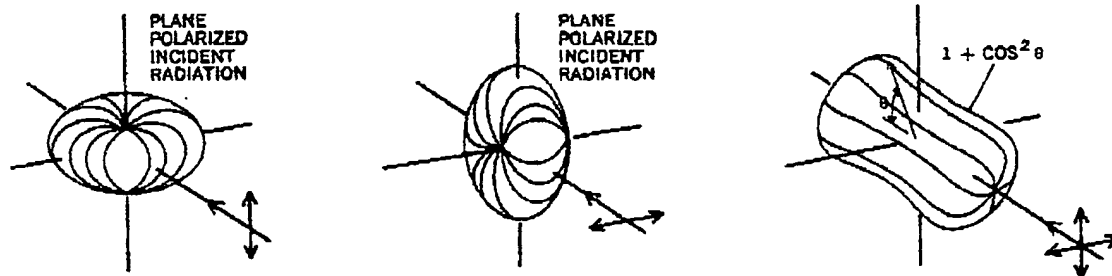
$$\tau l = -\ln \frac{I}{I_0} \quad [40]$$

can be written, for unit-cell length, as

$$\begin{aligned} \tau &= -\ln \left( \frac{I}{I_0} - \frac{I_0}{I_0} + 1 \right) \\ &= -\ln \left( \frac{I - I_0}{I_0} + 1 \right) \\ &\approx \frac{I_0 - I}{I_0} \end{aligned} \quad [41]$$

The term  $I_0 - I$  is the intensity removed from the incident beam and is therefore the integral over all angles of the scattered radiation of

**FIGURE 25-12**  
The angular dependence of secondary, or scattered, radiation from a particle that is small compared with the wavelength of the radiation.





Eq. [39]. The measure of turbidity can therefore be evaluated, according to the differential surface element of Fig. 25-13, as

$$\tau = \int_0^\pi \frac{i}{I_0} 2\pi r^2 \sin \theta \, d\theta \quad [42]$$

Substitution of the expression for scattered intensity  $i$  of Eq. [39] and integration gives

$$\tau = \frac{8\pi}{3} \left( \frac{2\pi}{\lambda} \right)^4 \alpha^2 \quad [43]$$

For a concentration of  $c$  g/cc and a molecular weight  $M$  there will be  $(c/M)\mathcal{N}$  molecules per cubic centimeter, and the turbidity of such a material will be

$$\tau = \frac{8\pi}{3} \left( \frac{2\pi}{\lambda} \right)^4 \alpha^2 \frac{c\mathcal{N}}{M} \quad [44]$$

To obtain a calculation of molecular weight from a measured turbidity, it is necessary to have a value of the molecular polarizability. The expression of Eq. [37] of Sec. 14-5 can be reduced for  $n_R \cong 1$ , as is the case for gases, to the relation between  $\alpha$  and the refractive index  $n_R$  of

$$2\pi \left( \frac{c}{M} \mathcal{N} \right) \alpha = n_R - 1 \quad [45]$$

Here  $c$  plays the role of  $\rho$ , used in Sec. 14-5. With this result the turbidity of a gaseous system, in which the particles are small compared with the wavelength, can be written as

$$\tau = \frac{32\pi^3 M (n_R - 1)^2}{3\lambda^4 c \mathcal{N}} \quad [46]$$

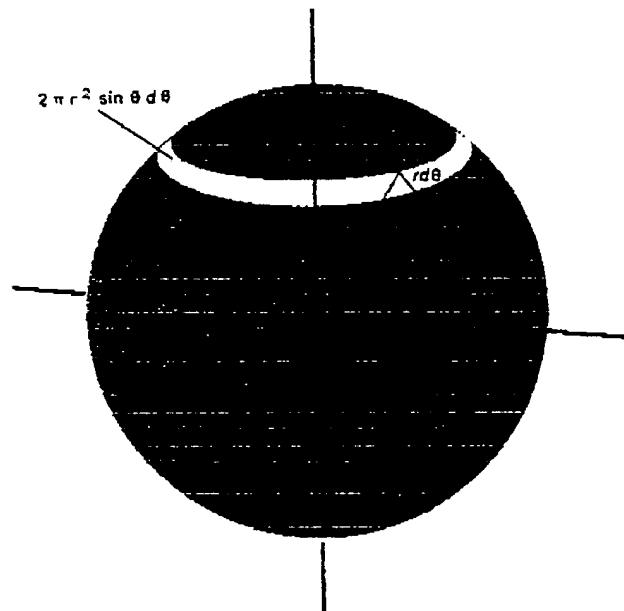


FIGURE 25-13

*The surface element for the integration of the scattered radiation.*

With this expression the measurable turbidity can be related to the molecular weight of the gas-phase particles, the mass of material per unit volume, and the refractive index of the gaseous system. This expression has, in fact, been turned around so that a value for Avogadro's number has been obtained from the scattering produced by gas samples.

For the systems of interest here, i.e., macromolecules in a liquid medium, it is necessary to introduce the fact that the scattering depends on the *difference* between the refractive index of the particles and that of the medium. If  $n_R$  is the refractive index of the solution and  $n_R^0$  that of the pure solvent, the appropriate relation comparable with Eq. [46] turns out to be

$$\tau = \frac{32\pi^3 M n_R^0}{3\lambda^4 c \mathcal{N}} (n_R - n_R^0)^2 \quad [47]$$

Now measurements of  $\tau$  and  $n_R$  for a solution of a given value of  $c$  and  $n_R^0$  allow the calculation of the molecular weight  $M$ . It should be mentioned that, since the polarizability increases with increasing molecular size, the amount scattered by an individual molecule is proportional to its size. The molecular weight that is obtained is therefore a weight-average molecular weight.

In practice, this expression is usually written as

$$\tau = \frac{32\pi^3 n_R^0}{3\lambda^4 \mathcal{N}} \left( \frac{n_R - n_R^0}{c} \right)^2 \frac{c}{M}$$

or

$$\tau = H \frac{c}{M} \quad [48]$$

where

$$H = \frac{32\pi^3 n_R^0}{3\lambda^4 \mathcal{N}} \left( \frac{n_R - n^0}{c} \right)^2 \quad [49]$$

In dilute solutions, moreover, the term for the change of refractive index with concentration can be written as a differential, and  $H$  then is

$$H = \frac{32\pi^3 n_R^0}{3\lambda^4 \mathcal{N}} \left( \frac{dn_R}{dc} \right)^2 \quad [50]$$

From measurements of refractive index for the wavelength of light used in the scattering experiments,  $H$  can be evaluated and the turbidity can be measured. Equation [50] can then be used to calculate a molecular weight. In practice, an extrapolation to infinite dilution is necessary, and, for particles of appreciable size compared with the wavelength of light, so also is recognition of the angular dependence of the scattered light.

A little must now be said about the scattering that results when the molecules are not small with respect to the wavelength of the light. Visible light has wavelengths between about 4000 and 8000 Å, and these lengths are just about the dimensions expected for many macro-

molecules. As for electron scattering from different atoms of a molecule in an electron-diffraction experiment, the scattering from different parts of the molecule will now interfere with one another. The effect is, in fact, very similar to that studied in detail for electron diffraction. The macromolecule is best thought of as some geometric shape presenting a continuum of scattering centers rather than a few discrete centers. A detailed calculation for the amount of light scattered as a function of angle, for a given wavelength and assumed molecular size and shape, can be performed by integrating the Wierl equation of Sec. 13-3 over all parts of the molecule. The type of light-scattering angular dependence that can result for molecules with dimensions like that of the wavelength of the scattered light is indicated in Fig. 25-14. The details of the pattern are dependent on the shape of the molecule as well as on its overall size. Measurements of light scattering now give promise of being one of the most powerful methods for studying the geometry of macromolecules in solution.

In lieu of a detailed analysis of the molecular shape that would lead to the observed angular dependence of the scattered radiation, it is often sufficient to measure the intensity of the scattered beam at two angles, usually  $45^\circ$  and  $135^\circ$ , to the incident beam. The ratio of these intensities reflects the overall shape of the macromolecule in solution. Calculations have been made, using essentially the Wierl equation for some simple shapes, and these are shown in Fig. 25-15. From observations of the scattered intensity at the angles of  $45^\circ$  and  $135^\circ$ , such curves can be consulted and lead to some information on the usually unapproachable quantity, the shape of a molecule in solution.

### 25-13. ELECTROKINETIC EFFECTS

The behavior of colloidal particles dispersed in an aqueous medium is greatly affected by the fact that the particles often carry an electric charge. The presence of acidic and basic groups in proteins, for example, means that there will generally be positive or negative charges on the protein molecule. The number and sign of these charges will depend on whether the solution is acidic or basic. Charges are also

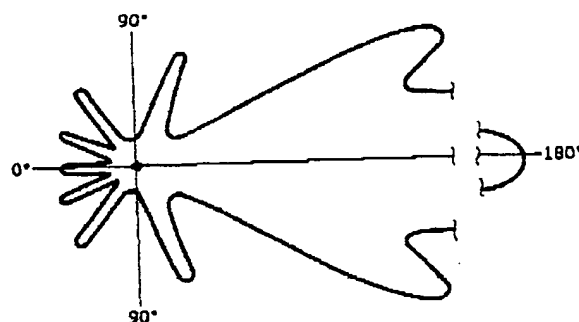


FIGURE 25-14

*The scattering of visible light from a spherical particle of radius 5000 Å. [From V. K. LaMer and M. Kerker, *Light Scattered by Particles*, Sci. American, 188:69 (1953).]*